Fine structure and differential metastability measurements for the doubly excited $1s 2s 2p {}^{4}P^{o}$ and $1s 2p {}^{2}{}^{4}P$ states of lithiumlike Ne VIII

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Fine-structure intervals have been measured for the $J = \frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$ levels of the doubly excited $1s 2s 2p {}^{4}P^{0}$, and $1s 2p {}^{2}{}^{4}P$ terms of lithiumlike Ne VIII, using fast-ion-beam spectroscopy in the far uv. In addition, lifetimes for the $J = \frac{1}{2}$ and $\frac{3}{2}$ levels of the upper term were determined, and their comparison with theory shows the importance of choosing for such levels atomic models which include magnetic interactions. Our measurement for the $J = \frac{3}{2}$ level of $1s 2p^{2}{}^{4}P$ is in excellent agreement with a recent relativistic calculation and removes a previous discrepancy between measured and theoretical values for this level.

I. INTRODUCTION

Three-electron atomic systems can occur in very highlying metastable states in which all spins are aligned. A quarter century ago unclassified lines emanating from lithium atoms in a hollow cathode source¹ were later interpreted as transitions between such doubly excited states. Since then, much activity has been centered on such states both in neutral lithium as well as in higher members of the isoelectronic sequence.² Most of this work has used foil-excited beams as a light source, as they tend to produce these states rather copiously.

Improvements in experimental techniques and increasingly sophisticated calculations have led to a recent flurry of activity in the study of the lowest-lying quartet states of Li-like systems, $1s 2s 2p {}^{4}P^{o}$ and $1s 2p^{2}{}^{4}P$. On the experimental side, improved resolution has led to determination of fine-structure intervals for both terms as well as the possibility of studying the differential metastability of the upper term. Levels of different J value will have different lifetimes, since both radiative and autoionization transition rates are sensitive to J. Data from a wide range of members of the lithium isoelectronic sequence provide information on spin-orbit, spin-other-orbit, and spin-spin interactions through their varying dependence on nuclear charge Z. Relativistic effects have been shown³ to play a very important role in determining decay rates for such states both for radiative and nonradiative processes.

The most recent calculations⁴ with which our lifetime and fine-structure data will be compared are carried out in relativistic intermediate coupling and are expected to be the most reliable to date. In fact, one of our principal justifications in studying Ne VIII in particular was the existence of a discrepancy of a factor of more than 2 between the calculated decay rate⁴ of the $J = \frac{3}{2}$ level of $1s 2p^{24}P$ in Ne VIII and the corresponding decay rate as measured by an Auger electron-emission experiment.⁵ The $J = \frac{1}{2}$ and $\frac{3}{2}$ states are sensitive to details of the atomic model, and in particular to the contribution of the magnetic interaction to the Auger decay rate.⁴ This is not so for the $J = \frac{5}{2}$ state where Auger decay takes place predominantly via mixing with doublet states. For this state, there is agreement between even nonrelativistic results⁶ and experiments on lower members of the lithium sequence.⁷

Previous measurements in this field beyond neutral lithium involved fine-structure and lifetime determinations for C IV, N V, and O VI.⁷ Fine structure for the F VII system was studied by Martinson *et al.*,⁸ who also obtained a lifetime for the $1s2p^{24}P_{5/2}$ level. Beyond the previously mentioned Auger-spectroscopy work in neon,⁵ there have been fine-structure studies in Mg X (Ref. 9) and Al XI,^{10,11} and a lifetime measurement for ${}^{4}P_{1/2}$ in Al XI.¹¹ Since submission of this manuscript, a parallel work¹² has been published on Ne VIII whose results are in good agreement with ours.

II. EXPERIMENT

All measurements were performed at 13 MeV using doubly charged beams of neon ions from a 7 MV Van de Graaff accelerator. A modified rf ion source¹³ furnished beam intensities in excess of 1 μ A (absolute particle current). Thin homemade self-supporting carbon foils served to excite the beam. Their thicknesses ranged between 36 and 48 μ g/cm² and were measured by comparing their stopping power (relative to air) for 5.5 MeV α particles.¹⁴ This resulted in an uncertainty of less than 1% in the beam velocity (10.9 mm/ns).

Photons emitted by the spontaneously decaying foilexcited ions and emitted in a direction perpendicular to the beam were dispersed by a 0.5 m McPherson Seya-Namioka monochromator equipped with a concave, masked, MgF₂-coated aluminum 600 groove/mm grating. Entrance and exit slits were set to 50 μ m, and the monochromator was refocused¹⁵ by increasing the exit slit-tograting separation by an amount appropriate for the beam velocity used. In this way, the reduced linewidths (~1 Å full width at half maximum) allowed resolution of the ${}^{4Po}{}^{-4P}$ multiplet into its fine-structure components (Fig. 1). Photons were counted with a Channeltron (Galileo).

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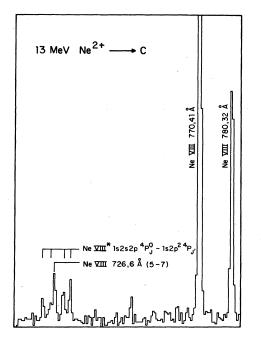


FIG. 1. Spectrum of foil-excited 13 MeV neon showing the resonance lines and transitions between low-lying ${}^{4}P$ states.

At least three scans were taken in the 450 to 800 Å region in order to calibrate the lines of interest (see Fig. 1). Wavelengths for three of the narrowest lines in the quartet multiplet ~726 Å were thus established (723.42, 729.67, and 731.70 Å). Well-established narrow Ne VII and Ne VIII lines¹⁶ were used for wavelength calibration: those at 465.221, 770.409, and 780.324 Å. The line positions were reproducible to within 0.05 Å. A slow scan of the region of interest (700–750 Å) was then performed at high resolution and the three lines mentioned above in this region were then used as "transfer standards" in the high-resolution scan. We estimate an overall uncertainty of ± 0.15 Å in our wavelength determinations.

A section of the beam ~ 1.2 mm in length is viewed by the optical system. At the stated beam velocity, this corresponds to a "time window" of about 0.1 ns (the instrumental function is in fact trapezoidal). Lifetimes were measured by displacing the foil target towards the accelerator and by observing the rate of decay of emitted light as a function of distance downstream from the foil. The latter was advanced by a screw drive and steppingmotor combination controlled by a Digital Equipment Corporation PDP-15 computer which also served to record data on-line. As beams of Ne²⁺ remain very stable over long periods, time elapsed per point and not accumulated beam charge was used to normalize photon counts per channel. Very rarely was it necessary to discard a decay curve due to a target perforated by the beam during a measurement.

III. DATA REDUCTION AND ANALYSIS

A. Wavelengths

As mentioned above, the positions of three lines in the region of interest (\sim 726 Å) were first placed on a firm footing with respect to known¹⁶ lines in the neon spectrum (465.221, 770.409, and 780.324 Å). These are at 723.42, 729.67, and 731.70 Å and are shown below to belong to the $1s 2s 2p {}^{4}P^{o} - 1s 2p^{2} {}^{4}P$ multiplet. A slow, high-resolution scan in the region of interest yielded additional lines at 721.00, 725.35, 726.6, and 727.15 Å. Since not all components could be resolved even after improvement of spectrometer resolution as described above, it was decided to extract blended components by deconvolution. A simple yet effective method,¹⁷ useful when only lowerorder moments of the convolution function are known, was applied to our data. The deconvoluted spectrum was then fitted by Gaussian profiles using the European Organization for Nuclear Research (CERN) program MINUIT.¹⁸ This procedure extracted a feature at 727.15 Å, which is presumed to contain the $\frac{5}{2} - \frac{5}{2}$ and $\frac{1}{2} - \frac{1}{2}$ finestructure components, based on theoretical predictions and experimental isoelectronic comparisons. We then assumed the positions of the $\frac{5}{2}$ - $\frac{5}{2}$ and $\frac{1}{2}$ - $\frac{1}{2}$ components to be at 727.0 and 727.3 Å, based on theoretical finestructure intervals listed in Hata and Grant.¹⁹ Line identifications were based partly on calculated values for transition energies and fine-structure intervals^{4,19-21} and part-

Fine-structure	Measured wavelengths (Å)		Predicted wavelengths (Å)			
components (J-J)	This work	Livingston <i>et al.</i> (Ref. 12) (±0.05 Å)	Cheng <i>et al.</i> (Ref. 20)	Chen <i>et al.</i> (Ref. 4)	Hata and Grant (Ref. 19)	Chung (Ref. 21)
$\frac{3}{2} - \frac{5}{2}$	721.00±0.2	721.12	714.1	726.3	720.1	720.79
$\frac{1}{2} - \frac{3}{2}$	723.42 ± 0.15	723.27	716.3	728.6	722.3	723.09
$\frac{3}{2} - \frac{3}{2}$	725.35 ± 0.2	725.60	718.2	730.7	724.5	725.14
$\frac{5}{2} - \frac{5}{2}$	727.15±0.2	(726.90)	720.1	732.6	726.2	726.87
$\frac{1}{2} - \frac{1}{2}$		(727.19)	720.4	732.9	726.5	727.29
$\frac{3}{2} - \frac{1}{2}$	729.67 ± 0.15	729.55	722.4	735.0	728.6	729.39
$\frac{5}{2} - \frac{3}{2}$	$731.70 {\pm} 0.15$	731.45	724.2	737.0	730.6	731.28
Center of gravity	726.71	726.51	719.5	732.0	725.5	726.36

TABLE I. Wavelengths of measured and predicted fine-structure components of the $1s 2s 2p {}^{4}P^{o}-1s 2p^{2} {}^{4}P$ multiplet in Ne VIII.

Interval	This work (±60)	Livingston <i>et al.</i> (Ref. 12) (±15)	Cheng <i>et al.</i> (Ref. 20)	Chen <i>et al.</i> (Ref. 4)	Hata and Grant (Ref. 19)	Chung (Ref. 21)
1 s 2s 2p ⁴ P ^o						· · · · · · · · · · · · · · · · · · ·
$\frac{1}{2} - \frac{5}{2}$	1564	1546	1525	1558	1577	1554
$\frac{1}{2} - \frac{3}{2}$	368	444	371	389	398	394
$\frac{3}{2} - \frac{5}{2}$	1196	1102	1154	1169	1179	1160
$1 s 2 p^{24} P$						
$\frac{1 s 2 p^{24} P}{\frac{1}{2} - \frac{5}{2}}$	1648	1602	1593	1626	1633	1637
$\frac{1}{2} - \frac{3}{2}$	816	746	804	810	812	805
$\frac{3}{2} - \frac{5}{2}$	832	856	789	816	821	832

TABLE II. Fine-structure splitting (in cm⁻¹) for the $1s 2s 2p {}^{4}P^{o}$ and $1s 2p {}^{2}{}^{4}P$ terms in NeVIII.

ly on isoelectronic comparisons with experimental work on other members of the lithium sequence.⁷⁻¹¹ We summarize these findings for the quartet states in Table I.

The analysis of this region was greatly complicated by the presence of a hydrogenic transition manifold $(n=5\rightarrow7)$ in NeVIII. Lindgård and Nielsen²² predict 5f-7g at 726.4 Å, 5d-7f at 726.7 Å, and 5g-7h at 726.8 Å. This last transition, the yrast transition of the manifold, is expected to be by far the strongest, based on many previous beam-foil studies. We indeed observe a rather broad feature at 726.6±0.2 Å which is accounted for by these transitions.

A more serious difficulty arises from the predicted²² positions of the transitions 5f-7d at 731.9 Å and 5p-7d at 729.4 Å. If these features were indeed found at the predicted positions they would mask two of the quartet lines in Table I. This is highly unlikely as the calculated²² lifetime is 0.0441 ns for the upper 7d state, and no such short-lived component was found in our lifetime measurements either at 729.7 or 731.7 Å.

The fine-structure intervals are listed in Table II and compared with the various available theoretical values and with a recent measurement. Agreement is excellent throughout, although the experimental uncertainty does not permit one to single out any one of the calculations as being closer to experiment than another.

B. Lifetimes

In the upper-state lifetime measurements, the decay curve corresponding to each transition used was repeated four or five times. The average values are listed in Table III. In particular, the decay curves representing the $J = \frac{3}{2}$ upper-state lifetime were taken via two transitions (at 723.4 and 731.7 Å), giving excellent agreement. The principal source of error is one of fitting multiexponentials to the decay curves. Beam-energy uncertainty is a mere $\pm 0.5\%$ due mainly to uncertainty in target-foil thickness.

The well-established program DISCRETE²³ served to analyze all the decay curves. The program has the advantage of requiring neither specification of starting values for the parameters, nor the number of exponentials present in the curve. In addition, it often provides a "second-best solution" where the most likely numerical fit is less than highly probable. In nearly all curves, the best solution consisted of a single exponential, with no cascading, and a constant background of the order of a few percent of the initial intensity of the primary decay. The constant background was consistent with that deduced from the spectra, and from a graphical analysis which was applied to all curves. The latter analysis gave excellent confirmation in all cases of the results of the numerical analysis generated by DISCRETE.

Our data for the lifetime work on the $1s 2p^{24}P$ term is represented in a condensed graphic form in Fig. 2. One can observe therein the excellent agreement between theory and experiment for the $J = \frac{1}{2}$ and $\frac{5}{2}$ states throughout the isoelectronic sequence. Previous confrontations between measured and calculated results were far less satisfactory for the $J = \frac{3}{2}$ level, however. The single previous datum in NeVIII for this level (using Auger electron-emission measurements), while in apparent agree-

TABLE III. Measured and theoretical lifetimes of the $J = \frac{1}{2}$ and $J = \frac{3}{2}$ levels of $1s 2p^{24}P$ in NeVIII.

			Theoretical lifetime (ns)		
- ·	Measured 1		Tunnel and Bhalla	Chen et al.	
Level	Present data	Other work	(Ref. 24)	(Ref. 4)	
$1s 2p^{24}P_{1/2}$	$0.53 {\pm} 0.05$	0.5 ± 0.05	0.53	0.53	
$1s 2p^{24}P_{3/2}$	0.40 ± 0.04	(Ref. 12) 0.19 ± 0.09	0.11	0.45	
1 0,2		(Ref. 5)			
		0.408 ± 0.04			
		(Ref. 12)			

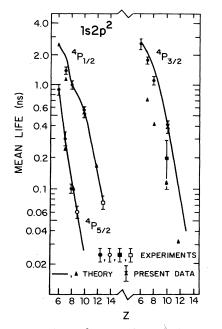


FIG. 2. Comparison of recent theoretical and experimental results for lifetimes of the $J = \frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$ levels of the $1s 2p^{24}P$ term in NeVIII. (The data of Ref. 12 coincide with ours.)

ment with a nonrelativistic calculation,²⁴ is in fact lower than the latest relativistic theoretical value⁴ by a factor of more than 2 (see also Table III). Our measurement for the $J = \frac{3}{2}$ level, on the other hand, confirms the recent calcu-

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- ¹G. Herzberg and H. R. Moore, Can. J. Phys. 37, 1293 (1959).
- ²See, for example, K. X. To, E. J. Knystautas, R. Drouin, and H. G. Berry, J. Phys. (Paris) Colloq. **40**, C1-1 (1979) and references therein.
- ³M. H. Chen, B. Crasemann, and H. Mark, Phys. Rev. A 24, 1852 (1981).
- ⁴M. H. Chen, B. Crasemann, and H. Mark, Phys. Rev. A 26, 1441 (1982).
- ⁵S. Schumann, K. O. Groeneveld, G. Nolte, and B. Fricke, Z. Phys. A **289**, 245 (1979).
- ⁶See, for example, F. Bely-Dubau, J. Dubau, P. Faucher, and L. Steenman-Clark, J. Phys. B **14**, 3313 (1981) and references therein.
- ⁷A. E. Livingston and H. G. Berry, Phys. Rev. A 17, 1966 (1978).
- ⁸I. Martinson, B. Denne, J. O. Ekberg, L. Engström, S. Huldt, C. Jupén, U. Itzén, S. Mannervik, and A. Trigueiros, Phys. Scr. 27, 201 (1983).
- ⁹E. Träbert, H. Hellmann, P. H. Heckmann, S. Bashkin, H. Klein, and J. D. Silver, Phys. Lett. **93A**, 76 (1982).
- ¹⁰E. Träbert, H. Hellmann, and P. H. Heckmann, Z. Phys. A **313**, 373 (1983).
- ¹¹J. P. Buchet, M. C. Buchet-Poulizac, A. Denis, J. Désesquelles, M. Druetta, S. Martin, J. P. Grandin, D. Hen-

lated value⁴ and thus shows the importance of including magnetic interactions in atomic models describing these systems. The recent results of Livingston *et al.*¹² support this conclusion.

IV. CONCLUSIONS

Measurements have been carried out on the doubly excited quartet states $1s 2s 2p {}^{4}P^{o}$ and $1s 2p {}^{2}{}^{4}P$ in lithiumlike Ne VIII. Fine structure for these states appears wellestablished, as measured values confirm recent calculated ones for all intervals. Furthermore, lifetime measurements on two J values of the upper state remove a previous discrepancy between theory and experiment and confirm the importance of magnetic interactions in atomic models used to calculate such states.

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- necart, X. Husson, and D. Lecler, J. Phys. (Paris) Lett. 45, L361 (1984).
- ¹²A. E. Livingston, J. E. Hardis, and H. G. Berry, Phys. Rev. A **30**, 2089 (1984).
- ¹³E. J. Knystautas, IEEE Trans. Nucl. Sci. NS-26, 1420 (1979).
- ¹⁴C. Jacques, thèse de maitrise, Université Laval, 1978 (unpublished).
- ¹⁵J. O. Stoner, Jr. and J. A. Leavitt, Appl. Phys. Lett. 18, 477 (1971).
- ¹⁶R. L. Kelly and L. J. Palumbo, Atomic and ion emission lines below 2000 Angstroms—hydrogen through krypton, U.S. Naval Research Laboratory (Washington, D.C.) Report No. 7599, 1973 (unpublished).
- ¹⁷E. Sjøntoft, Nucl. Instrum. Methods **206**, 199 (1983).
- ¹⁸MINUIT, CERN Program Library (Genève, Switzerland) (unpublished).
- ¹⁹J. Hata and I. P. Grant. J. Phys. B 16, 915 (1983).
- ²⁰K. T. Cheng, J. P. Desclaux, and Y.-K. Kim, J. Phys. B 11, L359 (1978).
- ²¹K. T. Chung, Phys. Rev. A **29**, 682 (1984) and private communication.
- ²²A. Lingård and S. E. Nielsen, At. Data Nucl. Data Tables 19, 533 (1977).
- ²³S. W. Provencher, Biophys. J. 16, 27 (1976); J. Chem. Phys.
 64, 2772 (1976); private communication.
- ²⁴T. W. Tunnel and C. P. Bhalla, Phys. Lett. 67A, 119 (1978).